Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020101

FWP and possible subtask under FWP:
FWP Dynamics and Structure of Interfaces and Dislocations
Subtask on Surface Dynamics

FWP Number: SCW604

Program Scope:
The goal of this project is to quantify the fundamental atomic processes governing the dynamics of surface structure and morphology. We use state-of-the-art microscopy (low-energy electron microscopy and scanning tunneling microscopy) to measure, often in real time, the time evolution of surface structure on nanometer length scales. We use these measurements to write down precise equations of motion to describe the observed time dependence and then relate these equations of motion to atomic processes. We have used this general approach on a variety of different problems in surface science. This work has often revealed unanticipated mechanisms of surface evolution. We currently emphasize four focus areas: 1) film wetting and de-wetting, 2) mass exchange between the bulk and surface, 3) surface self-assembly, and 4) oxide surfaces and metal oxidation. The goal of future work is to apply our approach to increasingly complicated material systems and further develop the conceptual framework needed to account for our observations. The ultimate goal is to provide the groundwork for quantitative predictions needed to engineer surface properties.

Major Program Achievements (over duration of support):
Key accomplishments include: determining the atomic-scale dynamics governing self-assembly of nanoscale patterns on surfaces; real-time observations and atomistic interpretation of the evolution of thin-film microstructure (e.g., twin-boundary motion); quantitative measurements of dislocation dynamics in thin film and their relationships to surface morphology, chemical reactivity and alloying; measurement of the thermodynamic stability of supported oxide nanostructures; discovery of new fundamental atomic mechanisms for the dynamics of surface morphology. In particular, we have determined the quantitative link between bulk vacancy formation and transport to surface smoothing and the role of substrate atomic steps in the de-wetting of thin metal films.

Program Impact:
By a combination of experiment and theory, we are among the first to be able to account quantitatively for the kinetic processes that determine the nanoscale surface structure. There is considerable recent interest in the materials science of nanoscale features on surfaces because of their possible applications in the development of new energy technologies. For example, understanding bulk-surface exchange is important for the development of hydrogen storage materials, and metal oxide surfaces are important industrial catalysts.

Interactions:
J. de la Figuera (Univ. Autónoma de Madrid); R. Q. Hwang (BNL); P. Hou, Andreas Schmid (LBNL); C. B. Carter (Univ. Minnesota); S. Chiang (UC Davis); J. B. Hannon (IBM Research); G. L. Kellogg, P. J Feibelman, B. S. Swartzentruber (SNL/NM); F. Besenbacher (Aarhus).

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
22 Publications (2003-2005), including one article in Science and 6 articles in Physical Review Letters. 2001 MRS Medal (Bartelt); 2001 Nottingham Prize (Thayer). Editorial boards of Physical Review Letter*s* and Surface Science (Bartelt) and Materials & Engineering Reports (McCarty).

Personnel Commitments for FY2005 to Nearest +/- 10%:
N. C. Bartelt (100%), K. F. McCarty (70%), K. Thuermer (70%), J. C. Hamilton (25%), J. P. Pierce (post-doc) (100%).

Authorized Budget (BA) for FY03, FY04, FY2005:
FY03 BA $1099k ($750k/349k) **FY04 BA $**1119k ($770k/349k) **FY05 BA $**1150k
Note: in FY05, FWP SCW1550 ("Oxide Surfaces and their Interactions with Metals") was combined with SCW604 subtask on "Surface Dynamics". The above budget numbers show the distribution between these two projects.

Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020101

FWP and possible subtask under FWP:
FWP Dynamics and Structure of Interfaces and Dislocations
Subtask on: Metallic Interfaces and Dislocations

FWP Number: SCW604

Program Scope:
This sub-program is concerned with establishing the basic principles that control the structure and behavior of internal interfaces. We concentrate on metallic interfaces, including both grain boundaries and heterophase interfaces, with a central goal of determining how the incompatibilities and discontinuities that arise at such interfaces are accommodated and what the implications of these relaxations are on the behavior and properties of the interface. One key to establishing this connection is in understanding the nature of interfacial defects. Just as bulk crystal behavior is dominated by the properties of lattice defects, one anticipates that interfacial processes are similarly controlled by the properties of point (e.g., vacancies and impurities) and extended defects (steps, dislocations, and junctions) present on the interface. A second key is in establishing how these defects interact with each other and with the interface itself. For instance, in real polycrystalline solids, boundaries of finite length terminate at junctions with other boundaries, and even a single boundary often deviates from the simple planar ideal, forming an array of atomistic-scale steps or breaking up into an array of larger scale facets. Finally, it is important to understand how interfaces are controlled by composition. We closely integrate both experiment and theory. Detailed experimental observations using high-resolution transmission electron microscopy, scanning tunneling microscopy, and atom-probe tomography on carefully controlled materials systems are combined with theory and computation encompassing continuum elasticity, interfacial crystallography, atomistic simulations, and first principles simulations. Our work continues to evolve from its early emphasis on determining the structure of simple and ideal planar grain boundaries to explaining the origin of more complex three-dimensional interfacial structures. Throughout, our approach is to employ both experimental and theoretical tools to obtain a basic scientific understanding of the fundamental structural elements, interactions, and excitations that govern interfacial behavior. Ultimately, a comprehensive picture of how these seemingly disparate structural elements work in concert will improve our ability to predict and control a diverse range of interface mediated materials processes including slip localization and transmission at boundaries, recrystallization and grain coarsening, grain boundary sliding, interfacial segregation and diffusion, and interfacial phase transformations.

Major Program Achievements (over duration of support):
Key accomplishments include: Discovery of structural phase transition for the {112} twin boundary; Quantitative analysis of size effects on the structure of grain boundary; Discovery of grain boundary interactions through extended stacking fault formation; Develop model explaining cluster shapes; Analysis of defaceting grain boundary transition and discovery of new faceting mechanism.

Program Impact: Have developed a quantitative understanding based on coupling simulation and microscopy of grain boundary structures.

Interactions:
C.B. Carter, University of Minnesota; Uli Dahmen, NCEM/LBL; Y. Mishin, George Mason University; I. Daruka, University of Debrecen, Hungary; A. Voter, LANL; S.M. Foiles, SNL/NM; R.C. Pond, Univ. of Liverpool, UK.

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
D.L. Medlin: Chair 2006 Gordon Research Conference on Physical Metallurgy.
J.C. Hamilton: Organizer of symposium at the Fall 2005 Meeting of the MRS

Personnel Commitments for FY2005 to Nearest +/- 10%:
D.L. Medlin (30%), J.C. Hamilton (40%), E. Marquis (50%)

Authorized Budget (BA) for FY03, FY04, FY2005:
FY03 BA $385k **FY04 BA** $385k **FY05 BA** $368k

Laboratory Name: Sandia National Laboratories, CA
B&R Code: KC020101

FWP and possible subtask under FWP:
FWP Dynamics and Structure of Interfaces and Dislocations
Subtask on: Alloying at Surfaces and Interfaces

FWP Number: SCW604

Program Scope:
The Alloying at Surfaces and Interfaces program at Sandia, California is focused on the development, validation and application of computational methods that can be used to explain and predict structural, mechanical, and thermodynamic properties of materials. We use and develop first-principles atomic scale and continuum methods. A current focus of the program is on using these techniques to understand the mechanisms of alloying and de-alloying. The ultimate goal of the program is to uncover fundamental concepts that can be applied to understanding the structure, behavior and processing of alloys. The main research areas are: (i) atomistic mechanisms that govern ultrathin film and surface alloy formation (ii) role of dislocations during phase separation of bulk alloys, (iii) role of H in energetics and kinetics of alloy formation and segregation, and (v) free energy in bulk and surface alloys.

Major Program Achievements (over duration of support):
Key accomplishments include: Development of continuum models to study the influence of dislocations on phase separation in binary alloys; Role of activation energies for surface and bulk intermixing in causing composition gradients during heteroepitaxy of Pd on Ru(0001) and Ge on Si(001); Effect of H on the formation of Al-Ti surface alloys and of vacancies in Ni; Development of anisotropic elasticity approach for self-assembly at solid surfaces, providing theoretical explanation for the unusual orientation of stripes in the PbCu system; Development of the self-consistent GW method, and applications to a broad range of materials; Development of first principles tool to determine vibrational entropies and test on H containing materials.

Program Impact: Has expanded our understanding of the mechanisms of solute stability in alloys and their implications to alloy behavior, and our understanding of the mechanisms of alloying and de-alloying.

Interactions:
Dr. Mikko Haataja (Princeton Materials Institute)
Dr. B. S. Swartzentruber (Sandia National Laboratories, Albuquerque, New Mexico)
Dr. J. B. Hannon (IBM T.J. Watson Research Center, New York)
Dr. J. J. Hoyt (Sandia National Laboratories, Albuquerque, New Mexico)
Dr. John Hamilton (Sandia National Laboratories)
Dr. Norman Bartelt (Sandia National Laboratories)
Dr. Andreas Schmidt (Lawrence Berkeley National Laboratories)
Dr. Nicholas Rougemaille (Lawrence Berkeley National Laboratories)

Recognitions, Honors and Awards (at least in some part attributable to support under this program):
R. Stumpf was co-organizer of a symposium on advanced H storage materials at the Fall 2004 MRS meeting. F. Leonard invited speaker and panelist at Nanocommerce conference, and part of advisory committee for joint chem/semi research for National Nanotechnology Initiative.

Personnel Commitments for FY2005 to Nearest +/- 10%:
R. Stumpf (50%), F. Léonard(50%), S. Faleev (50%) (post-doc)

Authorized Budget (BA) for FY03, FY04, FY2005:
FY03 BA $340k **FY04 BA $**349k **FY05 BA $**333k
Note: Formerly this task was entitled "Alloy Theory" and was under FWP SCW4341. It was combined with SCW604 in FY05.